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Submitted to:

*Physical Review Letters*  
August 2001



**Los Alamos**  
NATIONAL LABORATORY



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# Measurements of ultra-cold neutron lifetimes in solid deuterium

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(August 30, 2001)

## Abstract

Superthermal ultra-cold neutron sources offer orders of magnitude improvement in the available densities of ultra-cold neutrons, and are therefore of great importance to fundamental particle physics experiments such as searches

for a static electron dipole moment and lifetime measurements for the free neutron. We present the first measurements of the survival time of ultra-cold neutrons in solid deuterium. This critical parameter provides a fundamental limitation to the effectiveness of superthermal ultra-cold neutron sources which utilize solid ortho-deuterium as the source material. Our measurements are performed utilizing a solid-deuterium source coupled to a spallation source of neutrons, providing a demonstration of ultra-cold neutron production in this geometry and permitting systematic studies of the influence of thermal upscatter and contamination with para-deuterium on the UCN survival time. Valid PACS appear here. `\pacs{}` should always be input, even if empty.

*Introduction* Fermi first showed that the interaction of slow neutrons with the atomic nuclei in a material can be treated with an effective potential or index of refraction if the neutron wave length is long compared with the inter-atomic spacing. This potential,  $V$ , given by  $V = \frac{2\pi\hbar^2}{m}Na$ , where  $m$  is the neutron mass,  $N$  is the number density of the material, and  $a$  is the neutron scattering length, is as large as 335 neV for  $^{58}\text{Ni}$ . This corresponds to the kinetic energy of a neutron with a velocity of 8 m/s. Although Fermi and Zinn [1] experimentally demonstrated the total reflection of neutrons from the effective potential at material surfaces in 1946, Zel'dovich [2] in 1959 was the first to suggest that this effect could be used to bottle neutrons with kinetic energies below the effective potential of the bottle walls. He also recognized, that a sufficient number of slow neutrons might be obtained from the tail of the Maxwellian distribution near a reactor core to demonstrate this effect. Following this suggestion the existence of these neutrons was first demonstrated independently at the Petersburg Nuclear Physics Institute [3] and the Institut Laue-Langevin [4]. Neutrons with kinetic energies low enough to be trapped in material bottles are referred to as ultra cold neutrons (UCN) because of their extremely low kinetic energies of hundreds of neV. UCN densities at reactor sources have gradually increased with reactor power and improved techniques for extracting the UCN flux. The highest densities reported in the literature have been obtained at the ILL reactor in Grenoble [5].

Recent measurements of the neutron electric dipole moment [6,7] and the neutron lifetime (see, e.g. [8–10] attest to the utility of bottled UCN for fundamental experiments with neutrons. UCN may also be useful in improved measurements of angular correlations in neutron beta-decay, although experiments of this kind utilizing UCN have not yet been performed. All of these experimental programs have been limited by the available densities of UCN. It is in this context that we have performed measurements concerning superthermal UCN production in solid deuterium.

Superthermal UCN production was first proposed in 1975 by Golub and Pendlebury [11] and experimentally investigated shortly thereafter [12,13]. The concept is that phonon creation in a solid can be used to down-scatter cold neutrons to the ultra-cold regime while

up-scatter from phonons can be reduced to a negligible level by cooling the solid deuterium lattice. Solid deuterium is an ideal medium for this process because of its low absorption cross-section, low mass and relatively large coupling to neutrons. Superthermal production of UCN in superfluid He has been directly observed, and agrees reasonably well with theoretical expectations [10,13–16]. Recent efforts to utilize solid deuterium sources at reactors fell short of expectations, however, requiring more detailed investigation [12].

The limiting UCN density,  $\rho_{UCN}$ , one can obtain with a solid deuterium superthermal source is given by the product of the rate of UCN production in the solid,  $R$ , and the lifetime of UCN in the solid,  $\tau_{SD}$ :  $\rho_{UCN} = R\tau_{SD}$ . A larger storage bottle opened to such a source will come into density equilibrium with the density in the solid. This led to the proposal of a thin film source where the inside of a neutron bottle is coated with a thin layer of solid deuterium and is embedded in a cold neutron flux [17,18]. The volume comes into equilibrium with the UCN density with a time constant for the coupled system,  $\tau$ , given by,

$$\tau = \tau_{SD} \frac{V}{V_{SD}}, \quad (1)$$

where  $V_{SD}$  is the volume of solid deuterium in the source and  $V$  is the total volume of the storage bottle. A valuable summary of solid deuterium thin film sources is presented in [19]. The effects of gravity and of the potential of the solid as well as UCN losses other than absorption in the film have been neglected in this expression, but do not alter the above picture in a fundamental fashion. Ultimately, the limit to the UCN density is established by the trade-off between the cold neutron flux intensity and energy distribution (which determine the production rate) and the heat produced by neutrons and gamma rays in the solid deuterium and bottle walls, since  $\tau_{SD}$  is a strong function of temperature. However, the predicted production rates in solid deuterium [18] and lifetimes [20] have not been quantitatively verified yet.

A new source of UCN was suggested [21,22] that appears to solve the background problems associated with reactor sources and that may result in new UCN facilities with higher densities that have been obtained at reactors. The proposal was to drive a solid deuterium

superthermal converter with cold neutrons obtained by moderating spallation neutrons produced in a heavy target by a medium energy pulsed proton beam. In a spallation target the amount of heating for each neutron is lower than in a reactor, allowing higher neutron densities in the vicinity of a spallation target than can be achieved with a reactor. In addition, by pulsing the proton beam and valving off the UCN storage volume from the production volume when the beam is off, one can take advantage of these high neutron densities by using the time when the beam is off to remove heat from the deuterium. In this case the maximum UCN density one can produce is limited only by the impulse heating of the solid deuterium. Experiments with the stored UCN can be performed while the beam is off, eliminating the backgrounds due to capture gammas found near continuously operated reactor sources. In a similar way, a solid deuterium source could also be used at a TRIGA reactor, as it was proposed in [23]. We note that a Yoshiki *et al.* [24] attempted to couple a superthermal He source to a spallation target, with limited success. Although UCN were observed, the rates were not adequate to accurately assess whether the production rates and UCN lifetimes were in agreement with theoretical predictions.

In order to test the idea of a solid deuterium based spallation UCN source, we have built a test source that has been operated with single pulses of protons produced by the LANSCE 800 MeV proton accelerator at Los Alamos National Laboratory. In this letter we report the first measurements of  $\tau_{SD}$ , the lifetime of UCN in solid deuterium. Our measurements clearly indicate the critical influences of heating and para-deuterium contamination on the UCN lifetime, and provide a quantitative foundation for the development of a solid deuterium superthermal source at either a reactor or a spallation source.

*Experiment* A schematic view of our apparatus is shown in Fig. 1. Spallation neutrons were produced in a tungsten target with short (typically less than 160 ns long) pulses of 800 MeV protons from the LANSCE accelerator in Los Alamos. The fast neutron flux was contained and amplified using (n,2n) reactions in a layer of beryllium surrounding the spallation target. The spallation neutrons were moderated and cooled in a thin layer of polyethylene surrounding a  $^{58}\text{Ni}$  coated stainless steel guide tube with an inner diameter of

7.5 cm. The polyethylene and the bottom of the guide were cooled with liquid helium, and a layer of deuterium was frozen on the inside of it. UCN produced inside of solid deuterium are then confined by the guide tube, and could be directed through a series of valves to our UCN detector. Data were acquired using a multi-scalar that was started with a pulse produced by the proton beam passing through a toroidal pick-up coil and scaled the count rate from the UCN detector.

Several effects limit the lifetime for UCN in solid deuterium: upscatter from phonons in the solid [18], upscatter off of para-deuterium molecules in the solid [20,25], absorption on deuterium, and absorption on hydrogen impurities in the solid. Model calculations exist for the contribution of all of these effects on the UCN lifetime in the solid. One can write the total loss rate as the sum of contributions from each of the sources listed above:  $1/\tau_{SD} = 1/\tau_{phonon} + 1/\tau_{para} + 1/\tau_{D_{2abs}} + 1/\tau_{H_{abs}}$ , with the loss rate due to phonon upscatter having different contributions from the ortho- and para- deuterium in the solid. Establishing the experimental basis to validate these models as both accurate and complete is essential for the design of a UCN source based on the superthermal production mechanism in solid ortho-deuterium.

Solid deuterium was frozen in the lower part of the cryostat using a liquid helium refrigerator. The temperatures of the lower guide walls and of the liquid helium cryostat were monitored with an array of silicon diodes mounted on the guide and the aluminum cryostat walls. The temperatures of the thermometers tracked the vapor pressure curve of solid deuterium well at higher temperatures. The temperature of the solid at lower temperatures was obtained by averaging the temperatures of two diodes mounted on the outside of the guide wall. Later measurements made with diodes embedded in frozen solid indicate these measurements are accurate to 0.5 K.

Both the hydrogen contamination and the para fraction in the solid deuterium were measured by means of rotational Raman spectroscopy on a gaseous sample taken at warming up the deuterium after the measurement [20]. These measurements yielded values for the HD concentrations in the gas which varied across a range of 0.2-0.3% with an uncertainty

of about 0.1%. Other contaminations were removed by a palladium membrane in the  $D_2$  gas system, prior to introducing  $D_2$  to the cryostat. The para-fraction was controlled by converting the deuterium to a near thermal equilibrium ortho-para ratio in an iron hydroxide filled cell cooled to a temperature at or slightly below the triple point. In this way the para fraction was reduced from room temperature equilibrium value of 33% to 2-4%. Intermediate values were obtained by mixing deuterium at room temperature equilibrium with converted deuterium before freezing. The precision of the para-fraction measurements was typically of order of 1%.

The solid volume was measured by integrating the flow of gas while growing the solid. The solid volume was then calculated from the room temperature volume which was frozen. When the solid was warmed and the gas was returned to a buffer volume and its volume was checked. The uncertainties in the pressure dependence of calibration of the flow meter (20%), and uncertainties in the guide volume and temperature combined to lead to an uncertainty in the solid volume of about 20%.

Neutrons were detected in a 5 cm thick multiwire chamber detector filled with a mixture of  $^3\text{He}$  at 5 mbar and  $\text{CF}_4$  as a stopping gas at 1 bar. The low  $^3\text{He}$  pressure and the large bend angle in the guide resulted in a high degree of selectivity for detecting UCN in the apparatus. This selectivity was verified by measuring neutron arrival times with a 0.024 cm thick aluminum foil with and layer of  $^{58}\text{Ni}$  evaporated on its surface open and closed at valve location C. These data are shown in Fig. 2. The number of counts measured with the foil closed was 0.043 of the counts measured with the foil opened. About half of these could be attributed to UCN leakage through the gap around the outside of the valve, and the rest were neutrons with normal velocities sufficient to penetrate the potential barrier provided by the  $^{58}\text{Ni}$ . These data unambiguously demonstrate that the signal in the  $^3\text{He}$  detector at the end of our guide was predominantly from UCN for the lifetime measurements we report here.

If gravity, wall losses, the solid deuterium potential, and transport effects are neglected, and if the solid deuterium is thin enough so that its volume is uniformly sampled by the



neutrons, the lifetime of neutrons stored in a bottle in contact with solid deuterium is given by equation (1). We have used this idea to measure the life times of UCN in solid deuterium. As depicted in Fig. 1, valves B and C were open for these measurements. UCN were stored in the bottle between the end of the guide and valve A, which includes the solid deuterium. Figure 3 shows the number of UCN counts per beam proton and per  $\text{cm}^3$  of solid deuterium, as a function of the time when valve A was opened after the proton beam pulse. Exponential fits to a function of the form  $c_0 e^{-t/\tau}$ , where  $c_0$  and  $\tau$  were varied as described below to produce the best fit for the bottle lifetimes  $\tau$  for various conditions of the solid deuterium. Note that the background was independently determined by fitting the signal outside of the detection window to a linear function and did not contribute significantly to the statistical uncertainty of the extracted lifetime. The UCN lifetime in solid deuterium are extracted using the Monte Carlo analysis presented below. Data sets for different volumes of solid deuterium, at various temperatures and for various para/ortho fractions were obtained and analyzed.

*Data analysis and Results* A Monte Carlo program has been used to simulate the UCN interactions in the  $\text{SD}_2$  and transport to the detector. The full experimental geometry, gravitation, the  $\text{SD}_2$  potential (about 108 neV), and wall collisions in the guide tube have been taken into account. There are a number of parameters which need to be determined from our experimental data: the probability of UCN loss and non-specular reflection for each collision with the guide wall, the solid deuterium elastic scattering length and lifetime, and the physical configuration of the solid deuterium frozen on the walls (solid deuterium can freeze as a flat “pancake” on the bottom of the guide, or coat the entire He-cooled surface at the bottom the guide, in a “bucket”-shaped configuration).

Because we used very thin samples of solid deuterium for most of the lifetime measurements (0.4 to 4 cm), we were quite insensitive to the scattering length for UCN in solid deuterium. We can put a lower limit on the scattering length of roughly 4 cm from the scaling of direct UCN production measurements with  $\text{SD}_2$  volume, in which we observed roughly linear scaling of the UCN production rate with thickness of the solid deuterium

sample to quite thick samples (8 cm [26]). However, our extracted lifetime results change by less than 20 percent for pathlengths as small as .5 cm. We therefore used the theoretical value of 8 cm for the elastic scattering length for all of the results presented here.

The ratio of diffuse to specular reflections in wall collisions was adjusted to fit time-of-arrival spectra (see Fig. 2). The fraction of wall losses per wall collision and the shape of the solid deuterium ice were adjusted in a combined fit to reproduce the volume dependence for all of the low temperature data. The extracted values for the guide parameters (.975 for the ratio of diffuse to specular reflections) and wall losses of  $(2.0 \times 10^{-3})$  were consistent with those extracted from an independent set of measurements of guide transmissions and holding times at the Institut Laue-Langevin. The lifetimes due to absorption on deuterium and hydrogen were calculated using the known amount of hydrogen contamination, the tabulated thermal cross sections and the  $1/v$  dependence of the cross sections. The lifetimes in solid ortho and para deuterium due to temperature upscattering were taken from the literature as well [18,20]. The lifetime in solid para deuterium due to molecular transitions was treated as a free parameter and found to be  $\tau_{SD}^{para} = 1.2 \pm 0.2$  ms is consistent with the calculation of [20] which gives  $\tau_{SD}^{para} = 1.5$  ms.

Results for lifetimes  $\tau_{SD}$  of UCN in solid deuterium as a function of the  $SD_2$  temperature and para/ortho fractions are shown in Fig. 4. It is clear this figure that the effect of deuterium vapor in the guide has to be included for higher temperatures. With this caveat, the measured lifetimes agree well with theoretical predictions of the upscatter rate. The main contributions to the UCN lifetime in solid deuterium have been measured and are quantitatively understood. These data demonstrate that UCN sources based on solid deuterium converters can provide the promised large gains over existing sources.

*Acknowledgments* We would like to acknowledge the help of the LANSCE operations staff for their excellent job of beam delivery in the new pulsed modes needed for this experiment. We also acknowledge fruitful conversation and inspiration from R. Golub. This work has been supported by the DOE XK3XAKJ and the NSF 9420470, 9600202, 9807133.

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## FIGURES

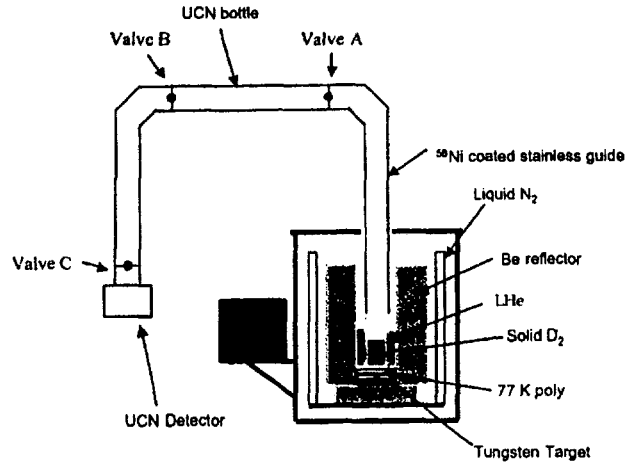


FIG. 1. Schematic view of the apparatus used for this experiment. Neutrons were bottled in the region between valves A and B. Valve C was used to insert a thin  $^{58}\text{Ni}$  foil coated aluminum foil in the guide before the detector. Solid lifetime measurements were made by measuring the number of UCN that survive in contact with the deuterium as a function of time using valve A, with valve B and C opened.

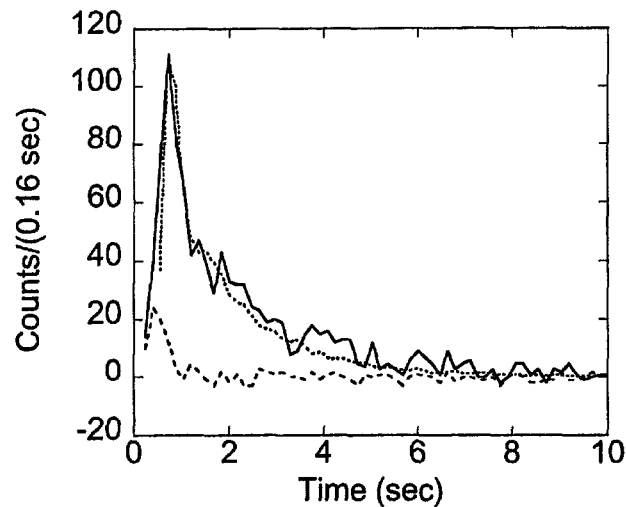


FIG. 2. Background subtracted spectra with and without the  $^{58}\text{Ni}$  barrier in the beam at the location of valve C. The dashed curve is the result of a Monte Carlo calculation of UCN arrival times.

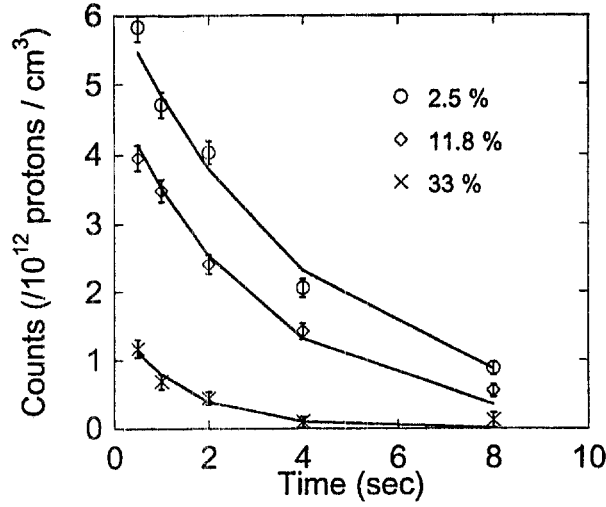


FIG. 3. Data and constrained exponential fits (solid lines) to bottlelife time data taken with different para-deuterium fractions.

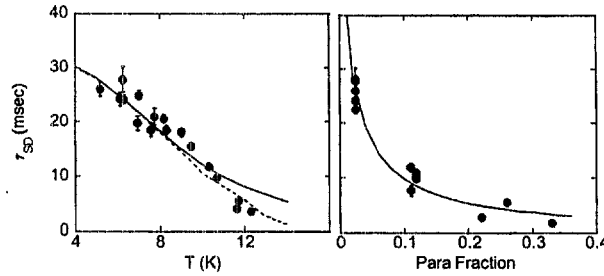


FIG. 4. Data points are fitted solid deuterium lifetimes as a function of temperature, para-fraction and volume. Solid lines show the predicted temperature and para-fraction dependence to the lifetimes. Dashed line is the predicted effect of departure from the solid lifetime model to up-scatter from the gas in the guide at higher temperatures. Solid lifetimes have been calculated by matching the measured bottle lifetimes with UCN in contact with the solid formed using valve A to a function of bottle lifetime vs. SD<sub>2</sub> lifetime generated using Monte Carlo. The SD<sub>2</sub> Geometry and the cold wall losses were adjusted in the Monte Carlo to fit the volume dependence.